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IONIZATION CLUSTER SIZE DISTRIBUTION FOR ALPHA PARTICLES: EXPERIMENT, MODELLING

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The paper presents data for measured ionization cluster size distributions by alpha particles in tissue equivalent media and comparison with the simulated data for liquid water. The experiments were carried out with a beam of 4.6 MeV alpha particles performed in a setup called the JET Counter. The theoretically derived cluster size distributions for alphas particles were obtained using the K-means algorithm. The simulation was carried out by Monte Carlo track structure calculations using cross sections for liquid water. The first moments of cluster size distributions, derived from K-means algorithm as a function of diameter of cluster centroid, were compared with the corresponding moments derived from the experiments for nitrogen and propane targets. It was found that the ratio of the first moments for water to gas targets correlates well with the corresponding ratio of the mean free paths for primary ionization by alpha particles in the two media. It is shown that the cluster size distributions for alpha particles in water, obtained from K-means algorithm, are in agreement with the corresponding distributions measured experimentally in nitrogen or propane gas targets of nanometer sizes.

INTRODUCTION

The aim of this work was to compare the calculated cluster size distribution for 4.6 MeV alpha particles in liquid water with the experimentally obtained data in nitrogen and propane as gaseous substitutes for tissue. The data provides a tool for consistency check of calculated clusters of ionizations at nanometer level in water and the corresponding experimental data in the gaseous media.

Ionizing radiation induces damage in genomic DNA in the form of simple and clustered damage. The hit region of DNA may contain strand breaks and base damages of various degrees of complexity. In the absence of direct experimental evidence, Monte Carlo track structure simulation and biophysical models have provided considerable understanding and quantitative information on the complexity of DNA damage, the types and the source of damage[(1–3)]. Theoretical modelling of clustered DNA damage formation by charged particles have been carried out by simulation and analysis of charged particle track formation in liquid water as a surrogate for the medium of the cell[(1–3)]. Experimental approach[(4)] have also become possible in the recent past. In the experiment the environment was replaced by a nanometer size gas target. Nitrogen and propane appeared to be the most convenient media for the experiments. Previously, it was shown[(5)] the possibility to obtain experimental data on the distribution of ionization clusters formed by charged particles within cylindrical volumes with sizes up to 1.3 μg cm^{-2} (13 nm in unit density scale). This paper compares the formation of clusters of ionizations by 4.6 MeV alpha particles in liquid water with the measured data in nitrogen and propane.

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MATERIAL AND METHODS

Theoretical

To provide an objective and quantitative measure of output sensitivity to variations of input parameters and model assumption, formal cluster analysis methods have been incorporated in the evaluation of the stochastics of charged particle tracks. The algorithm used in this work is K-means, a general computational tool for looking for correlation, meaning 'close' by distance, among different types of interactions along the track in a systematic fashion. Input requirements of the algorithm include an initial partition of the ensemble of data into K groups or clusters. In the simple algorithm used in these calculations, cluster number K is conserved, that is, the algorithm neither adds nor deletes clusters. Within the algorithm, the centroid of the data within each initial cluster is evaluated along with a measure of the discordance by attempting systematically to reassign a datum to every other cluster. The algorithm stops when no acceptable reassignment is found for one complete pass through the data ensemble. In this way the algorithm achieves a local minimum as opposed to a global minimum that would require testing of all possible permutations of the data, a problem too large for most powerful hardware.

As a measure of discordance of the data within a cluster, the algorithm uses the sum over the transfer points in the cluster of their squared distances from the centroid of the cluster (in the context of this paper, 'transfer points' are synonymous to

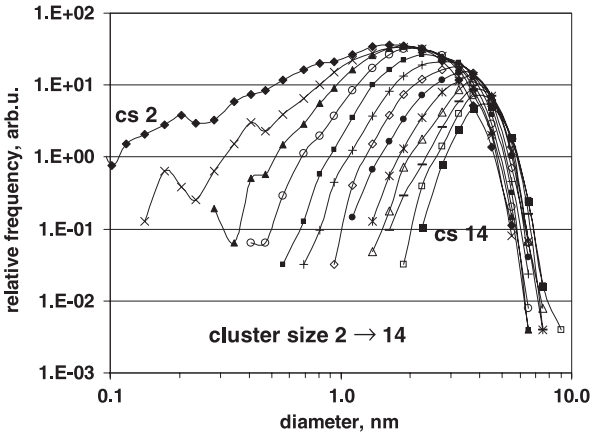

Figure 1. Frequency of ionization cluster size vs cluster diameter for clusters of 2–14 ionizations.

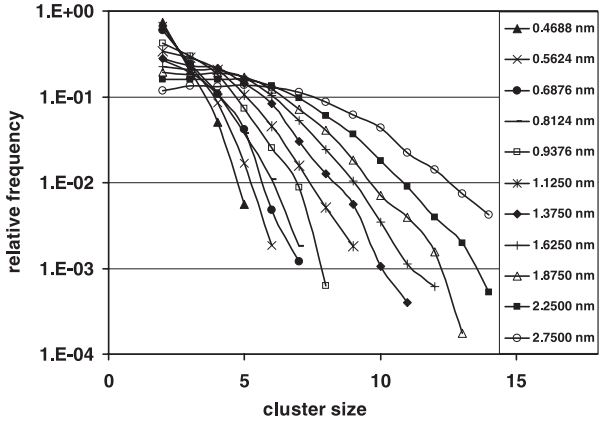

Figure 2. Frequency of ionization cluster size vs cluster size for different cluster diameter.

ionizations). This is a quantity akin to χ^2. By taking the square root of this quantity, finally returned by the algorithm, an *rms* measure of the physical radius of each cluster was returned. Also, summing the energy associated with each transfer point in the final clusters defines the total energy deposited in terms of a radiation-quality akin to linear energy transfer (LET). A large number of 1μm particle-track segments generated by the code PITS99[6] were analyzed. In this work we scored only those ionizations produced by the core primary ion track and the delta electrons of 100 eV or less. In this energy restricted LET sense, we are interested in the ionizations by the primary particle and delta electrons with lateral extension of a few nanometer from the primary ion path. For the initial partition, the coordinates of the ionizations in the track were sorted into ascending order along path of the ion. Cluster analysis were then carried out using K-Means algorithm[7]. Figure 1 shows frequency distribution of cluster size as a function of the cluster diameter for 4.6 MeV alpha particles. Figure 2 shows frequency distribution of clusters for different cluster sizes containing 2–14 ionizations.

Experimental - Modeling of nanometre size target

In this work it was assumed that ionization processes at nanometre level can be modeled in a gas cavity with an appropriate size. A gas cavity which simulates a nanometre size volume (in unit density) was created as shown in Figure 3. A simulated nanometre size target (SNS) was obtained by injecting a pulse of gas (nitrogen or propane) into the interaction chamber (IC). The gas density in the IC was controlled by the gas pressure in the reservoir R.

The collimated 4.6 MeV alpha particles from the source (gold plated AMM2 type Amersham)

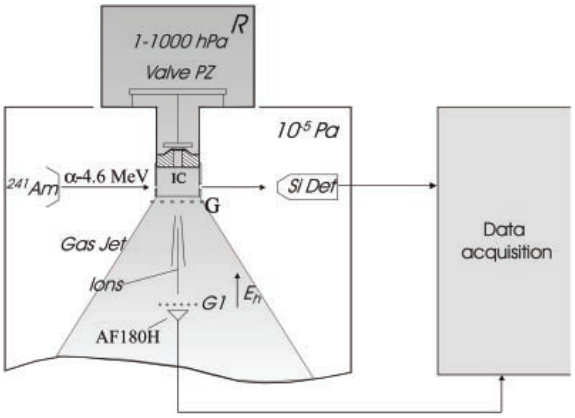

Figure 3. Schematic view of the Jet Counter: configuration for the experiments with alpha particles, PZ- piezoelectric valve; AF180H- ion detector, Si alpha particle detector, IC interaction chamber.

intersect the SNS chamber at half of its height and were registered by a Si detector. The ions created by a single alpha particle along its path within the SNS were removed by an electric field applied to the detector. The cluster size spectra in nitrogen and propane corresponding to different nanometre sizes ranging from 0.1 to 0.5 μg cm^{-2} were measured. The results are shown in Figure 4.

RESULTS AND DISCUSSION

It has commonly been accepted that nanometer size structures of two media can be assumed equivalent when the first moments of frequency distributions of cluster size versus cluster size have equal values. The first moment, $M_{1,E}$, of the measured cluster size distribution, P_ν (T) versus the cluster size ν, for alpha

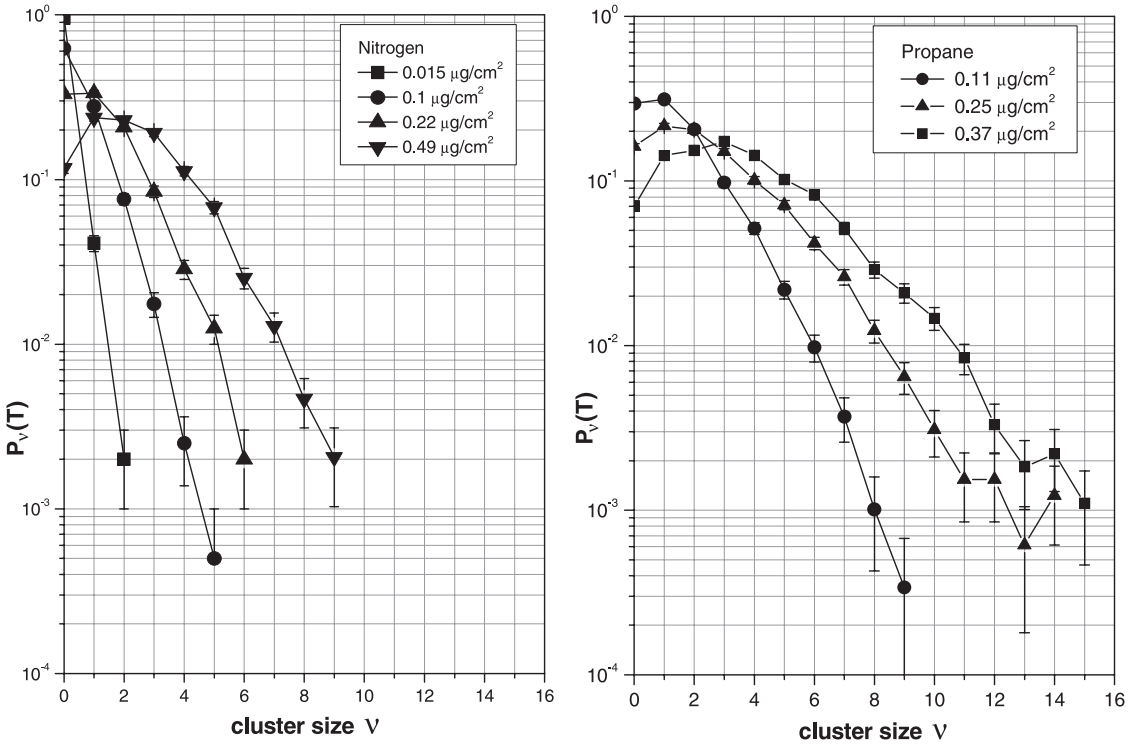

Figure 4. Experimental frequency distributions of ionization clusters for different target diameters for nitrogen and propane, irradiated by 4.6 MeV alpha particles. For propane the energy of alpha particles is attenuated by Mylar foil (1μg cm^{-2}).

particle of energy T was calculated as:

$$M_{1,E} = \sum_{\nu=0}^{\infty} \nu P_\nu(T); \text{assuming that} \sum_{\nu=0}^{\infty} P_\nu(T) = 1.$$

The first moment, M_1, for the experimental cluster size distributions for propane and nitrogen were calculated from the measured values, $M_{1,E}$, using the relationship $M_1 = M_{1,E}/\eta$, where η is the efficiency of single ion counting by the ion detector. Figure 5 shows the first moments of experimental (propane and nitrogen) and the calculated (water) cluster size distributions as a function of cluster diameter.

As seen from Figure 5, dependence between the first moment for water, propane and nitrogen can be expressed by a linear function of the cluster diameter with very small values of the intercept (within the experimental error it can be assumed that this dependence is proportional). On the other hand, as has been pointed by Grosswendt[8], for small cavity sizes i.e. for the case when delta electron contribution to cluster size yield can be neglected, the following relation exists, namely: $M_1 = d\rho/L\rho$, where dρ is the cluster diameter or diameter of the simulated nanometer size in mass per unit area, Lρ is the mean free path (*mfp*) for primary ionizations of alpha particles in the given medium in mass per unit area and ρ is the density of the gas. The results

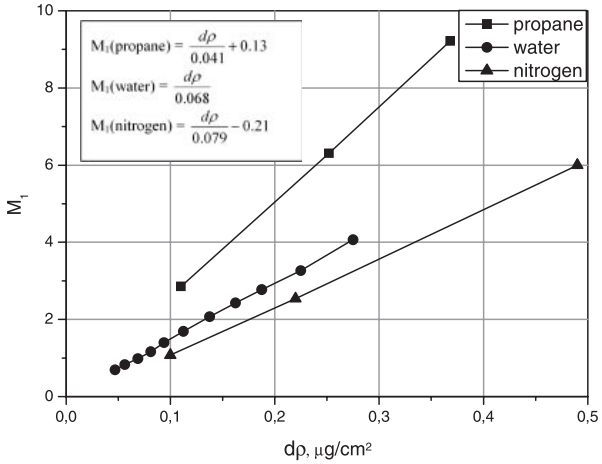

Figure 5. First moments M_1, of the calculated cluster size distribution for liquid water and the experimental cluster size distributions for nitrogen and propane.

presented in Figure 5 support this notion. The calculated *mfp* values for 4.6 MeV alpha particles are: 0.0637, 0.0511 and 0.0920 μg cm^{-2} for water, propane and nitrogen respectively. As seen from Figure 5 the slopes of M_1 for the investigated media follow the *mfp* data. Also, Table 1 shows the ratios of M_1 for water, propane and nitrogen for different cluster size diameters, are consistent with

Table 1. Ratio of M_1 for water, propane and nitrogen for different cluster diameters.

Cluster diameter (nm)	Water/ propane	Water/ nitrogen	Nitrogen/ propane
1	0.57	1.40	0.41
2	0.58	1.27	0.46
3	0.59	1.24	0.48
4	0.59	1.22	0.49

the *mfp* values. This consistency allows: (1) to compare cluster size distributions in water, as a surrogate for tissue like material, with those obtained in propane and nitrogen; (2) to scale cluster size distributions in nitrogen or propane to an equivalent cluster size distribution in water.

CONCLUSIONS

The first moments of cluster size distribution for water, derived from track structure simulation using K-means algorithm as a function of the diameter of cluster centroid, were compared with the corresponding moments derived from the experiments with nitrogen and propane targets. It was found that the ratio of first moments of cluster size distributions for water to nitrogen and propane gas targets correlates well with the corresponding ratios of mfp for primary ionizations by alpha particles in these media. It was shown that the cluster size distributions for alpha particles in water, obtained from K-means algorithm, are in agreement with the corresponding distributions measured experimentally in nitrogen or propane gas targets of nanometer sizes.

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